



# Coarsening versus pattern formation

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## Abstract

It is known that similar physical systems can reveal two quite different ways of behavior, either coarsening, which creates a uniform state or a large-scale structure, or formation of ordered or disordered patterns, which are never homogenized. We present a description of coarsening using simple basic models, the Allen-Cahn equation and the Cahn-Hilliard equation, and discuss the factors that may slow down and arrest the process of coarsening. Among them are pinning of domain walls on inhomogeneities, oscillatory tails of domain walls, nonlocal interactions, and others. Coarsening of pattern domains is also discussed.

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## 1. Introduction

For many decades, the nonlinear development of instabilities in physical systems was an object of extensive investigations. The most spectacular consequences of instabilities are the appearance of ordered spatially non-uniform structures (*pattern formation*, see [1] - [3]) or irregular motions (*spatio-temporal chaos* [4]) under uniform external conditions. However, there is one more scenario of an instability development: that is a gradual growth of the characteristic scale with time (*coarsening*) [5], [6]. Different evolution scenarios can take place in rather similar physical systems.

As an example, let us consider the phase separation in binary alloys that consist of two kinds of atoms, A and B, with volume fractions  $\phi_A(\mathbf{x}, t)$  and  $\phi_B(\mathbf{x}, t)$ , respectively. There exists a temperature  $T_c$  such that for  $T > T_c$  the components are mixed, i.e., the order parameter  $\phi(\mathbf{x}, t) = \phi_A(\mathbf{x}, t) - \phi_B(\mathbf{x}, t)$  vanishes everywhere, while for  $T < T_c$  they are separated, i.e., there exist two thermodynamically stable phases, one with  $\phi > 0$  (“A-rich phase”) and the other with  $\phi < 0$  (“B-rich phase”). A mathematical model of that phenomenon has been suggested by Cahn and Hilliard [7]. Under the simplest assumption of a constant mobility, the kinetics of the phase separation is described by the following non-dimensional equation (*the Cahn-Hilliard equation*),

$$\phi_t = \nabla^2(-\phi + \phi^3 - \nabla^2 \phi). \quad (1)$$

The uniform phase  $\phi = 0$  is unstable while the uniform phases  $\phi = \pm 1$  are stable. The instability of the phase  $\phi = 0$  creates a mosaic of islands of both stable phases. The size of these islands (*domains*) grows due to coarsening, which eventually leads to a complete separation of stable phases [5], [6].

A diblock copolymer, which consists of monomers A and B with reduced equal local densities  $\phi_A$  and  $\phi_B$ , is quite similar to a binary alloy. The basic difference is the existence of a long-range interaction of monomers [8] - [10]

which provides an additional term in the evolution equation for the order parameter:

$$\phi_t = \nabla^2(-\phi + \phi^3 - \nabla^2\phi) - \Gamma\phi, \quad \Gamma > 0, \quad \langle\phi\rangle = 0. \quad (2)$$

There are no other spatially uniform stationary solutions except  $\phi = 0$ . Therefore, when the latter solution is unstable (at  $\Gamma < 1/4$ ), a transition to a non-uniform state is unavoidable [11]. At small  $\Gamma$ , the initial evolution of disturbances is similar to that in the Cahn-Hilliard equation, but it is stopped when stripes with a definite pattern wavelength are created [12].

The existence of long-wave linear instability and multiple homogeneous states does not guarantee the creation of spatially uniform domains through coarsening. As an example, let us discuss the nonlinear dynamics governed by the one-dimensional Kuramoto-Sivashinsky equation,

$$\phi_t = -\phi_{xx} - \phi_{xxx} - (\phi^2)_x, \quad \langle\phi\rangle = 0, \quad (3)$$

which is used for the description of instabilities in reaction-diffusion systems [13], [14], instabilities of flame fronts [15], and film flow instabilities [16]. In that case, the linear part of the equation is identical to that of the Cahn-Hilliard equation, and any constant solution,  $\phi = \phi_0$ , satisfies equation (3). However, all solutions corresponding to uniform states are unstable. The Kuramoto-Sivashinsky equation is a paradigmatic model of the *spatio-temporal chaos* [14]; stable periodic patterns are also possible [16], [17], but the attraction domain of that regime is small. Generally, the way of the instability development depends significantly on the details of the system nonlinearity and symmetry [18], [19].

Note that pattern formation and coarsening are not incompatible phenomena. Let us return to model (2) that describes formation of stripes. Because of the rotational invariance of the problem, the orientation of stripes is arbitrary. Initially, a disordered system of stripes is developed from random initial conditions, and then the mean size of ordered domains grows with time, i.e., domain coarsening takes place for differently oriented stripe patterns rather than for different uniform phases [11], [20].

One can see that the interplay between coarsening and pattern formation is nontrivial, and it is the subject of the present chapter. Let us emphasize that here we discuss only *dynamic models* of coarsening and pattern formation, which do not include any kind of noise. The phenomena caused by thermal fluctuations are considered in other papers of the present issue and in the comprehensive book [21], where the reader can find many additional references on that subject.

## 2. Coarsening in one dimension: dynamics of domain walls

When considering the kinetics of coarsening, one has to take into account the following basic factors.

1. *The existence of a Lyapunov functional.* If the temporal evolution of an  $n$ -component order parameter  $\phi_i$ ,  $i = 1, \dots, n$  is described by a *gradient* evolution equation

$$\phi_{i,t} = - \sum_{k=1}^n D_{ik} \frac{\delta F}{\delta \phi_k}, \quad i = 1, \dots, n, \quad (4)$$

where  $F = \int L(\phi, \phi_x, \dots) dx$  is the Lyapunov functional of the system, and  $D_{ij}$  is a positive definite matrix, then

$$F_t = \sum_i \int \dot{\phi}_i \frac{\delta F}{\delta \phi_i} dx = - \sum_{i,k} \int D_{ik} \frac{\delta F}{\delta \phi_i} \frac{\delta F}{\delta \phi_k} dx \leq 0. \quad (5)$$

In the case of an equilibrium phase transition, the existence of the Lyapunov functional (free energy) is the consequence of the thermodynamics. The nonlinear development of instabilities in systems far from equilibrium in some cases is also governed, at least approximately, by *potential* systems of equations that possess Lyapunov functionals.

2. *The existence of a conservation law.* In the case where the order parameter is a density of a conserved quantity (e.g., the number of molecules), the evolution equations have a *divergence* form,

$$\phi_{i,t} = -\nabla \cdot \mathbf{J}_i, \quad i = 1, \dots, N, \quad (6)$$

where the flux  $\mathbf{J}_i$  is a function of the order parameter and its derivatives.

In order to understand how both factors influence the coarsening kinetics, let us consider a number of examples.

## 2.1. Non-conserved order parameter

### 2.1.1. Allen-Cahn equation

We start with the Allen-Cahn equation [22]

$$\phi_t = \phi_{xx} + \phi - \phi^3, \quad (7)$$

which describes a phase transition in the absence of a conservation law for the order parameter  $\phi$ . The physical interpretation of that model can be as follows [6]:  $\phi$  is the spontaneous magnetization directed along the definite axis (due to the crystal anisotropy). Also, the Allen-Cahn equation is the simplest example of an order parameter equation for a pattern forming system far from the thermodynamic equilibrium. Let us consider the onset of convection in a horizontal cylinder heated from below. Above the instability threshold, the rotation of the liquid in the transverse section of the cylinder can be either counterclockwise ( $\phi > 0$ ) or clockwise ( $\phi < 0$ ). The temporal evolution of the order parameter  $\phi(x, t)$  ( $x$  is the coordinate along the axis of the cylinder) is governed by the Allen-Cahn equation [23].

Equation (7) has a Lyapunov functional,

$$F(t) = \int L(x, t) dx, \quad L(x, t) = \frac{1}{2} \phi_x^2 + \frac{1}{4} (\phi^2 - 1)^2 \geq 0. \quad (8)$$

Its derivative

$$\dot{F} = \int \left( \frac{\partial L}{\partial \phi} \phi_t + \frac{\partial L}{\partial \phi_x} \phi_{xt} \right) dx = \int (-\phi + \phi^3 - \phi_{xx}) \phi_t dx = - \int dx \phi_t^2 \leq 0 \quad (9)$$

is non-positive, hence the Lyapunov functional decreases monotonically with time until a stationary state is reached. It is obvious that equation (7) has three fixed points corresponding to uniform phases,  $\phi = 0$  (unstable paramagnetic phase) and  $\phi = \pm 1$  (ferromagnetic phases with opposite orientations of the magnetization); the latter solutions correspond to the absolute minima of the Lyapunov functional,  $F = 0$ . The separatrices

$$\phi_{\pm} = \pm \tanh \frac{x - \xi}{\sqrt{2}}, \quad \xi = \text{const} \quad (10)$$

(*kink* and *antikink*) describe *domain walls* separating semi-infinite domains with different signs of the order parameter. Note that

$$\phi_{\pm} \sim \pm \{1 - 2 \exp[\mp(x - \xi) \sqrt{2}]\} \text{ as } x \rightarrow \pm\infty. \quad (11)$$

The contribution of a domain wall to the Lyapunov functional is  $F_0 = 2 \sqrt{2}/3 > 0$ .

Other stationary solutions of (7) describe *spatially periodic patterns* which can be considered as periodic arrays of domains with alternating signs of  $\phi$ . They are expressed through the elliptic Jacobi function.

Let us impose an infinitesimal disturbance  $\hat{\phi}(x, t)$  on the stationary solution  $\phi(x)$  and consider its temporal evolution governed by the linearized problem,

$$\hat{\phi}_t = \hat{\phi}_{xx} + (1 - 3\phi^2(x))\hat{\phi}, \quad |\hat{\phi}| < \infty \text{ as } x \rightarrow \pm\infty. \quad (12)$$

For normal disturbances  $\tilde{\phi}(x) \exp(\sigma t)$  we obtain an eigenvalue problem,

$$\sigma \tilde{\phi} = \tilde{\phi}'' + (1 - 3\phi^2)\tilde{\phi}, \quad |\tilde{\phi}(\pm\infty)| < \infty. \quad (13)$$

It is obvious that solution  $\phi = 0$  is unstable:  $\sigma(\tilde{k}) = 1 - \tilde{k}^2$  for  $\tilde{\phi} = e^{i\tilde{k}x}$ , and the solutions  $\phi = \pm 1$  are stable:  $\sigma(\tilde{k}) = -2 - \tilde{k}^2$  for the same  $\tilde{\phi}$ . The domain wall solutions (10) are neutrally stable. One can show that all the spatially-periodic solutions mentioned above are unstable.

Let us discuss now the temporal evolution of the system, when the initial state is the unstable phase  $\phi = 0$  with a certain initial, spatial disordered, perturbation. According to the stability analysis presented above, the final state should be uniform or contain a single defect, a domain wall. However, it is clear that the decomposition of the phase  $\phi = 0$  can produce numerous domains with alternating signs of  $\phi$ . Such a state can be characterized by a certain *density of defects* decreasing with time, or by a *mean domain length* which grows with time. At late stages of the

evolution, when the typical distance between domain walls is large the analysis can be done by means of asymptotic methods [24], [25]. One considers a set of domain walls (10) of alternating signs centered at  $\xi_i = \xi_i(t)$ ,  $i = 1, 2, \dots$ , and slowly moving because of their interaction. By means of asymptotic expansions, the original nonlinear problem is transformed into an infinite system of inhomogeneous linear equations. Their solvability conditions determine the following equations of motion for the centers of domain walls [26]:

$$\frac{2\sqrt{2}}{3}\dot{\xi}_i = -\frac{\partial U}{\partial \xi_i}, \quad (14)$$

where

$$U = \sum_i W(\xi_i - \xi_{i-1}), \quad W(\xi_i - \xi_{i-1}) = -8\sqrt{2} \exp[-\sqrt{2}(\xi_i - \xi_{i-1})]. \quad (15)$$

Thus, the domain walls attract to each other according to the exponential interaction law (14) which reflects the exponential domain wall asymptotics (11).

System (14) has a family of stationary solutions

$$\xi_j = a + jl, \quad j = 0, \pm 1, \pm 2, \dots \quad (16)$$

corresponding to periodic patterns with the spatial period  $2l$ . The attractive interaction makes all these solutions unstable.

Consider the interaction of a pair of domain walls. Setting  $l(t) = \xi_2(t) - \xi_1(t)$ , we find that the distance between domain walls is governed by the equation

$$\frac{2\sqrt{2}}{3}l'(t) = -32e^{-l\sqrt{2}}. \quad (17)$$

If  $l(0) = l_0 \gg 1$ , the solution is

$$l = l_0 + \frac{1}{\sqrt{2}} \ln \left( 1 - 48e^{-\sqrt{2}l_0 t} \right). \quad (18)$$

The distance between two domain walls becomes of  $O(1)$  at

$$t \sim t_0 = \frac{1}{48}e^{\sqrt{2}l_0} + O(1). \quad (19)$$

Finally, the domain walls reach the distance of order  $O(1)$  and annihilate. During the time interval  $t$ , only the domain walls with the original separation greater than  $\sim \ln(48t)/\sqrt{2}$  can survive. Thus, a logarithmic coarsening takes place. In a large but finite system with the length  $L$ , the number of domain walls  $N(t) \leq L/l(t)$ .

As an example of the situation when two locally stable phases are energetically non-equivalent, let us consider a system with the Lyapunov functional

$$F[\phi(x)] = \int \left[ \frac{1}{2}\phi_x^2 + \frac{1}{4}(1 - \phi^2)^2 - h\phi \right] dx \quad (20)$$

which corresponds to the dynamic equation

$$\phi_t = \phi_{xx} + \phi - \phi^3 + h. \quad (21)$$

In the case of a magnetic system, the last term in the expression (20) describes the influence of an external magnetic field, which makes the orientation of the magnetization in the direction of the field preferable. The uniform stationary states satisfy the equation

$$\phi - \phi^3 + h = 0. \quad (22)$$

In the interval  $-h_* < h < h_*$ , equation (22) has three solutions: stable solution  $\phi = \phi_+ > 1/\sqrt{3}$ , another stable solution  $\phi = \phi_- < -1/\sqrt{3}$ , and an intermediate unstable solution  $\phi_0$ ,  $-1/\sqrt{3} < \phi_0 < 1/\sqrt{3}$ . For  $h > 0$ , the phase with  $\phi = \phi_+ > 0$  is stable, and the phase with  $\phi = \phi_-$  is metastable.

If  $h$  is small, domain walls are described by formulas (10) at the leading order. By means of an asymptotic analysis, one can find that the motion of each domain wall is governed by the equation [26]

$$\frac{2\sqrt{2}}{3} \frac{d\xi}{dt} = \mp 2h \quad (23)$$

(the upper sign is for a kink and the lower sign is for an antikink). This motion creates a coarsening process, which is significantly faster than that in the case of energetically equivalent phases. That process leads to the elimination of the metastable phase.

### 2.1.2. Fractional Allen-Cahn equation

The logarithmic law of the one-dimensional coarsening is caused by the exponentially weak interaction, due to the exponential asymptotics (11) characteristic for solutions of partial differential equations. However, the basic equations governing the natural phenomena are often integro-differential equations rather than partial differential equations. For instance, the free energy density of a fluid depends on the fluid density in a nonlocal way [27]. The conventional van der Waals' expression for the fluid free energy which contains a squared density gradient [28] corresponds to a certain asymptotic limit of the basic nonlocal expression. The asymptotics of a domain wall (i.e., the gas-liquid boundary) in the framework of local and nonlocal models are strongly different: while a local model predicts an exponential decay, a nonlocal model suggests a power-law decay [27]. Fronts that cannot be governed by local partial differential equations have been found also in studies of transitions with long range interactions [29], vacancy diffusion and domain growth in binary alloys [30], and ordering kinetic on fractal structures [31] (for the latter subject, see [32]). As an example let us discuss the front propagation in systems with *superdiffusion* [33]. While the normal diffusion is associated with a Gaussian non-correlated random walk of particles, the superdiffusion is observed in non-equilibrium systems with an algebraically decaying jump length distribution, where the central limit theorem is no valid. Among the examples are wave turbulence [34], transport in porous media [35], and forage trajectories of animals [36]. One can use a superdiffusive generalization of the Allen-Cahn equation,

$$\phi_t = D_{|x|}^\gamma \phi + \phi - \phi^3, \quad 1 < \gamma < 2. \quad (24)$$

Here  $D_{|x|}^\gamma$  denotes the fractional Riesz derivative, which can be defined by its action in the Fourier space:

$$F\left(D_{|x|}^\gamma \phi(x)\right)(k) = -k^\gamma F(\phi(x))(k), \quad (25)$$

where  $F$  is the symbol of the Fourier transform. Equation (24) has a Lyapunov functional [37]. In a contradistinction to the exponential asymptotics (11) of the domain walls in the *local* PDE (7), the domain walls in the integro-differential equation (24) has an algebraic tail,

$$\phi_+ \sim 1 - \frac{\sec(\pi\gamma/2)}{2\Gamma(2-\gamma)} x^{-\gamma}, \quad x \gg 1. \quad (26)$$

That leads to a power law for the kink-antikink attraction,

$$l'(t) = -Cl^{-\gamma} \quad (27)$$

(cf. (17)), and for the temporal decay of the number of domain walls on a finite spatial interval,  $N \sim t^{-1/\gamma}$  [37].

### 2.1.3. Non-potential systems

For systems far from equilibrium, e.g., in the case of longwave instabilities of flows, the Lyapunov functional generally does not exist. Nevertheless, coarsening may take place if the domain walls are described by monotonic functions. As an example, let us mention the amplitude equation that governs the fixed-flux convection in a tilted slot [38]:

$$\phi_t = \phi_{xx} + \phi - \phi^3 + 2\alpha\phi\phi_x, \quad \alpha > 0. \quad (28)$$

Because the symmetry  $x \rightarrow -x$  is violated, the kink and antikink domain walls have different widths:

$$\phi_\pm(x) = \tanh \beta_\pm(x - \xi), \quad \beta_\pm = \frac{1}{2}(\alpha \pm \sqrt{\alpha^2 + 2}). \quad (29)$$

The interaction of domain walls is attractive but asymmetric. For a pair which consists of a kink with the center in the point  $\xi_1(t)$  and an antikink with the center in the point  $\xi_2(t)$ , the equations of motion look as

$$\xi_1' = F_+ \exp(-2|\beta_-|(\xi_2 - \xi_1)), \quad \xi_2' = F_- \exp(-2\beta_+(\xi_2 - \xi_1)), \quad (30)$$

where  $F_+ \neq F_-$ . All the periodic stationary solutions (patterns) are unstable, and a logarithmically slow coarsening takes place, similarly to the case of the Allen-Cahn dynamics.

## 2.2. Conserved order parameter

### 2.2.1. Potential systems

Let us return to the Cahn-Hilliard equation (1), which can be written as

$$\phi_t + j_x = 0, \quad j = (\phi_{xx} + \phi - \phi^3)_x; \quad -l \leq x \leq l. \quad (31)$$

Being a generic nonlinear equation governing longwave instabilities in the presence of the conservation law [18], that equation has been revealed in numerous problems of different physical nature, including secondary flows produced by the instability of the Kolmogorov flow [39], Marangoni instability of a two-layer system with a deformable interface [40], nonlinear development of zigzag instability of convection rolls [41], and even coarsening of ordered domains in oscillatory patterns governed by the complex Swift-Hohenberg equation [42], which describes oscillations in lasers [43], [44] and optical parametric oscillations [45] - [47].

For sake of simplicity, apply the boundary conditions  $\phi_x = \phi_{xxx} = 0$  at  $x = \pm l$ ; then

$$j(\pm l) = 0, \quad \frac{d}{dt} \int_{-l}^l \phi dx = 0 \quad (32)$$

(the total length of domains of each phase is conserved). The Lyapunov functional (8) decreases with time:

$$F_t = - \int (\phi_{xx} + \phi - \phi^3)^2 dx \leq 0. \quad (33)$$

Using the boundary conditions, one can present equation (31) in the form [25]

$$\partial_x^{-2} \phi_t + \phi_{xx} + \phi - \phi^3 + h(t) = 0, \quad \partial_x^{-2} \phi_t(x, t) = \frac{1}{2} \int_{-l}^l |x - y| \phi_t(y, t) dy. \quad (34)$$

Note that despite the energetical equivalence of phases  $\phi = \pm 1$ , an efficient field  $h(t)$  appears, which must to be determined self-consistently. The motion of a kink and an antikink towards each other would change the total lengths of domains of different phases, and hence it is not possible. Two kinks can move simultaneously towards an antikink placed between them, or kink-antikink pairs can move as a whole. The correlated motion of  $n$  domain walls with the centers at  $\xi_i$ ,  $i = 1, \dots, n$ , sufficiently far from each other, is governed by the system [25]

$$- \sum_{j=1}^n 2(-1)^{i-j} |\xi_i - \xi_j| \xi_j' = 16 \sum_{j \neq i} e^{-|\xi_i - \xi_j| \sqrt{2}} \text{sign}(\xi_j - \xi_i) + 2(-1)^i h(t), \quad i = 1, \dots, n, \quad (35)$$

supplemented by the conservation law

$$\sum_{i=1}^n (-1)^i \xi_i' = 0. \quad (36)$$

For a kink-antikink pair, the attraction is compensated by the field  $h = 8 \exp[-(\xi_2 - \xi_1) \sqrt{2}]$ , hence the domain walls are motionless. For a symmetric kink-antikink-kink triplet with coordinates of domain walls  $\xi_1 = -l(t)$ ,  $\xi_2 = 0$  and  $\xi_3 = l(t)$ , one obtains  $h = 0$ ,  $l' = -8 \exp(-l \sqrt{2})$ , hence the annihilation time depends on  $l_0 = l(0)$  as

$$t_0 \sim \frac{1}{8 \sqrt{2}} l_0 e^{l_0 \sqrt{2}}$$

(cf. (19)).

### 2.2.2. Non-potential systems

As an example of a non-potential system with a conservation law let us consider *the convective Cahn-Hilliard equation*,

$$\phi_t + (\phi_{xx} + \phi - \phi^3)_{xx} - \frac{D}{2}(\phi^2)_x = 0, \quad -\infty < x < \infty, \quad (37)$$

which has been suggested to describe several physical processes, namely spinodal decomposition of phase separating systems in an external field [53] - [55], step instability on a crystal surface [56], faceting of growing, thermodynamically unstable surfaces [57] - [61], evolving nanofoams [62] as well as dewetting of a thin film flowing down an inclined plane [63]. That equation provides “a bridge” between the Cahn-Hilliard equation (1) ( $D = 0$ ) and the Kuramoto-Sivashinsky equation (3) ( $\phi \rightarrow -2\phi/D$ ,  $D \gg 1$ ).

Stationary patterns  $\phi = \phi(x)$  are described by the problem

$$\phi''' + (\phi - \phi^3)' - \frac{D\phi^2}{2} = -\frac{DA}{2}, \quad -\infty < x < \infty; \quad A > 0; \quad x \rightarrow \pm\infty, \quad |\phi| < \infty. \quad (38)$$

For any  $D \neq 0$ , the set of solutions of equation (38) is incomparably more complex than that of the usual Cahn-Hilliard equation. One can easily find some exact solutions of the problem. The constant solutions,

$$\phi = \phi_{\pm} = \pm \sqrt{A}, \quad (39)$$

correspond to two stable phases. For domain walls, there exist *exact* solutions [53], one for a kink with  $A = A_+ = 1 + D/\sqrt{2}$ ,

$$\phi = \phi_+(x) = \phi_+^0 \tanh \frac{\phi_+^0}{\sqrt{2}}(x - \xi), \quad \phi_+^0 = \sqrt{1 + D/\sqrt{2}}, \quad \xi = \text{const}, \quad (40)$$

and the other for an antikink with  $A = A_- = 1 - D/\sqrt{2}$ ,  $D < \sqrt{2}$ ,

$$\phi = \phi_-(x) = -\phi_-^0 \tanh \frac{\phi_-^0}{\sqrt{2}}(x - \xi), \quad \phi_-^0 = \sqrt{1 - D/\sqrt{2}}, \quad \xi = \text{const}. \quad (41)$$

However, the set of stationary solutions is much more rich. Specifically, solution (40) is just one representative of a *family* of kinks  $\phi_+(x; A)$ . In addition to the monotonic antikink (41), there exists also a discrete set of non-monotonic antikink solutions [64] (that phenomenon is typical for models containing higher-order spatial derivatives, see [65]-[67]). The kink-antikink pair is formed by antikink (41) and a representative of the family of kinks with  $A = A_-$ .

If  $0 < D < D_0 = \sqrt{2}/3$ . In that region, the coarsening is observed [55], [58], [59], [61]. Because of the asymmetry between kinks and antikinks, a kink-antikink pair moves spontaneously with a definite velocity  $v_2(D, L)$ . The most typical process observed by coarsening is the annihilation of domain wall triplets, when two kinks of the same sign approach with velocities  $\pm v_3(D, L)$  the kink of the opposite sign situated between them. Exact expressions for  $v_2(D, L)$  and  $v_3(D, L)$  can be found in [68]. In the limit of small  $D$  [61],

$$v_2(D, L) \sim v_3(D, L) \sim -(D^2 \sqrt{2}/4) \exp(-DL/2).$$

Therefore, the coarsening law is logarithmic.

For  $D > D_0$ , the domain walls have oscillatory tails. That case will be discussed in the next section.

### 3. Factors hindering coarsening

In the present section, we discuss some typical situations where the system cannot reach a uniform state or another energetically preferred state by coarsening.

### 3.1. External inhomogeneities

The motion of domain walls leading to annihilation can be stopped by inhomogeneity of the medium. Recall that we consider the phenomena in the absence of noise. Coarsening in inhomogeneous systems in the presence of thermal fluctuations is considered in [69].

In a potential system, the domain wall would “prefer” the location where its energy will be smaller than in other locations. For example, let us consider the following modification of the one-dimensional Allen-Cahn equation [23]:

$$\phi_t = \phi_{xx} + [1 + \epsilon f(x)]\phi - \phi^3. \quad (42)$$

At the leading order in  $\epsilon$ , the equation of motion for a domain wall of any kind is

$$\frac{2\sqrt{2}}{3} \frac{d\xi}{dt} = -\frac{d}{d\xi} V_{ih}(\xi), \text{ where } V_{ih}(\xi) = \frac{1}{2} \int_{-\infty}^{\infty} f(\xi + y) \cosh^{-2}(y) dy. \quad (43)$$

Specifically, if the inhomogeneity has a  $\delta$ -like shape,

$$f(x) = -2V_0\delta(x - x_*),$$

the interaction potential is

$$V_{ih}(x_0) = -V_0 \cosh^{-2} \frac{x_0 - x_*}{\sqrt{2}}.$$

Generally, the shape of the potential is a linear transformation of the inhomogeneity shape, according to (43).

If there are many domain walls and many inhomogeneities, the motion of domain walls is determined by the system of equations (14) with the potential

$$U = \sum_i W(\xi_i - \xi_{i-1}) + \sum_i V_{ih}(x_i),$$

where  $W(\xi_i - \xi_{i-1})$  is determined by equation (15), and  $V_{ih}(x_i)$  corresponds to (43). Thus, the problem of finding stationary solutions of (42) is equivalent to finding equilibrium configurations of a chain of particles in the external potential (43), interacting according to the law (15). This model resembles the well-known Frenkel–Kontorova model (see e.g. [70]).

As an example let us consider two domain walls with coordinates  $\xi_1$  and  $\xi_2$  which are near the distant  $\delta$ -shaped attracting inhomogeneities:

$$f(x) = -2V_0\delta(x - x_{1*}) - 2V_0\delta(x - x_{2*}), \quad V_0 > 0, \quad x_{2*} > x_{1*}.$$

$x_{2*} - x_{1*} = l_* \gg 1$ ,  $|\xi_1 - x_{1*}| = O(1)$ ,  $|\xi_2 - x_{2*}| = O(1)$ . The equation of motion for the left domain wall is:

$$\frac{2\sqrt{2}}{3} \frac{d\xi_1}{dt} = 16e^{-(\xi_2 - \xi_1)\sqrt{2}} - \sqrt{2}V_0 \sinh \frac{\xi_1 - \xi_{1*}}{\sqrt{2}} \cosh^{-3} \frac{\xi_1 - x_{1*}}{\sqrt{2}}. \quad (44)$$

The first term in the right-hand side of equation can be estimated as  $16 \exp(-l_* \sqrt{2})$ . The minimum of the second term in the right-hand side of the equation is equal to  $-2\sqrt{2}V_0/3\sqrt{3}$ . Thus, we come to the conclusion that if

$$\frac{2\sqrt{2}}{3\sqrt{3}} V_0 > 16e^{-l_* \sqrt{2}}, \quad (45)$$

the domain wall will not be able to escape from the potential well created by the inhomogeneity. Hence, the coarsening will be stopped when the distances between the neighbor domain walls satisfy the inequality (45).

Similarly, in the case of domain walls pushed by the asymmetry of phases (see (23), we find the criterion of pinning:

$$V_0 > \frac{3\sqrt{3}}{\sqrt{2}} h.$$

If  $f(x)$  is a periodic function, the sequence of pinning sites (minima of the potential) filled by pinned domain walls can be regular (“commensurate patterns”) [71] or irregular (“spatial chaos”) [72].



### 3.2. Oscillatory tails of domain walls and stability of stationary patterns

In the examples considered in Sec. 2, the domain walls are described by monotonic functions like (10). The monotonicity of the asymptotic behavior of the domain wall solution on the infinity leads to a sign-preserving (attracting) interaction between domain walls. Oscillatory tails of domain walls create a sign-alternating interaction potential. The domain walls can be captured near the potential minima, therefore stable patterns are formed due to pinning of a domain wall by an inhomogeneity created by another domain wall.

As an example, let us consider the stability of periodic solutions of the convective Cahn-Hilliard equation (37), which satisfy the condition  $\phi(x + l) = \phi(x)$ . At large  $l$ , these solutions resemble periodic sequences of domain walls. Define the pattern wavenumber  $K = 2\pi/l$ . The normal disturbances of a periodic solution have the shape of a Floquet-Bloch function,  $\hat{\phi}(x, t) = \tilde{\phi}(x) \exp(ikx + \sigma t)$ , where  $\tilde{\phi}(x + l) = \tilde{\phi}(x)$ , and  $k$  is a quasi-wavenumber,  $|k| < K/2$ . A periodic solution is always neutrally stable ( $\sigma = 0$ ) with respect to a spatial shift,  $\tilde{\phi}(x) = \phi_x(x)$ ,  $k = 0$ . Therefore, a special attention has to be paid to potentially unstable longwave disturbances with small  $k$ . Their growth rate  $\sigma(k; K)$  can be presented as

$$\sigma(k; K) = \sigma_1(K)k + \sigma_2(K)k^2 + \dots$$

One can show that the sign of  $\sigma_1^2(K)$  depends on the dependence of the squared pattern amplitude

$$A = \frac{1}{l} \int_0^l \phi^2(x) dx$$

(see (38)) on the pattern wavenumber  $K = 2\pi/l$  [16], [73], [64]. If  $dA/dK < 0$  for any  $K$ , which takes place for  $D < D_0 = \sqrt{2}/3$ , then  $\sigma_1^2(K) > 0$  for any  $K$ , therefore all periodic solutions are unstable. That is compatible with the attractive interaction between domain walls. For  $D > D_0$ , the function  $A(K)$  is not monotonic, and the extrema of  $A(K)$  separate the regions of a monotonic growth of longwave disturbances,  $\sigma_1^2(K) > 0$ , and those of the oscillatory response of patterns to dilations and compressions,  $\sigma_1^2(K) < 0$  [74]. The region of oscillatory response can contain a subinterval of stable patterns (where  $\sigma_2(K) < 0$ ) [60], [64]. That is possible because of the alternating sign of the interaction between domain walls. On the boundaries of the stability interval, the patterns become unstable with respect to either longwave (phase) disturbances (see [75], [76]) or shortwave disturbances with  $k = K/2$ , leading to a spatial period doubling.

Because the potential of the interaction between domain walls has multiple minima, the distance between domain walls is not selected in a unique way. The pattern includes elements with “short” and “long” distances between the maxima that alternate in a rather irregular way [60].

### 3.3. Nonlocal interaction

A specific kind of an arrested coarsening process has been found for equation (2) [12], which can be written also as

$$\partial_x^{-2}(\phi_t + \Gamma\phi) + \phi_{xx} + \phi - \phi^3 + h(t) = 0. \quad (46)$$

For small  $\Gamma$  the lowest density of the Lyapunov functional is achieved for patterns with wavelength  $\lambda_{opt} = O(\Gamma^{-1/3})$ . According to the linear stability theory, the disturbance with largest growth rate has a wavelength  $\lambda_c = O(1)$ . Therefore, one could expect that the energetically preferable, long-wave pattern will be developed from the initial short-wave pattern by coarsening. The coarsening process takes place indeed, but it is stopped when the wavelength reaches a much smaller value,  $\lambda_{min} = O(\ln(1/\Gamma))$ . The criterion of the pattern stabilization is similar to (45), but now the stabilizing factor is the nonlocal interaction which is proportional to  $\Gamma$ .

### 3.4. Pattern-induced pinning of a domain wall.

The stability regions of a periodic pattern and a uniform state may overlap. In that case, the behavior of a *domain wall between the pattern and the uniform state* is crucial. Near the threshold of the instability creating short-wave patterns, where the width of a domain wall is large compared to the characteristic pattern wavelength, one can describe the dynamics of a domain wall using the envelope function approach [77], [78]. In the framework of that approach one comes to the conclusion that a domain wall between the pattern and the uniform state moves with a constant velocity, which is proportional to the difference between the Lyapunov functional densities of the phases [41]. However, the

influence of the underlying periodic pattern leads to some qualitative changes of the domain wall dynamics. First, the motion of the domain wall is an oscillatory process; during one period, one stripe is created or melted [79], [80]. Secondly, because of the pinning effect, there is a finite interval of the parameter value where the domain wall is motionless, i.e., a pattern and a uniform state coexist. Near the threshold of the pattern appearance, that interval is transcendently small [81], [41].

As an example, let us consider the competition and coexistence between patterns and uniform states for a system governed by the Swift-Hohenberg equation,

$$\phi_t = \left[ \epsilon - \left( \frac{\partial^2}{\partial x^2} + 1 \right)^2 \right] \phi - \phi^3 \quad (47)$$

which corresponds to the Lyapunov functional

$$F\{\phi\} = \int \left\{ -\frac{\epsilon}{2} \phi^2 + \frac{1}{4} \phi^4 + \frac{1}{2} \left[ \left( \frac{\partial^2}{\partial x^2} + 1 \right) \phi \right]^2 \right\} dx. \quad (48)$$

That model was suggested for studying hydrodynamics fluctuations near the instability threshold [82] and used for modelling Bénard convection [83]. At  $\epsilon > 0$ , periodic patterns exist with wavenumbers  $k$  in the interval  $1 - \sqrt{\epsilon} < k^2 < 1 + \sqrt{\epsilon}$ , and they are stable in a subinterval  $k_-(\epsilon) < k < k_+(\epsilon)$ . At  $\epsilon > 1$ , constant nonzero solutions  $\phi_{\pm} = \pm \sqrt{\epsilon - 1}$  appear. At  $\epsilon > 3/2$  they become stable with respect to small disturbances; kinks with oscillatory tails connect both stable uniform phases  $\phi_{\pm}$  [84].

The value of the Lyapunov functional density for the regular pattern with an optimal wavenumber is lower than that of the uniform state when  $\epsilon < \epsilon_m \approx 6.3$  [84], [79]. Nevertheless, the domain wall between both states is immobile for much smaller values of  $\epsilon$ ,  $\epsilon > \epsilon_c \approx 1.7574$ . The reason is the self-induced pinning caused by the oscillatory asymptotic perturbation of the uniform state. Similarly, the pinning effect prevents the replacement of a pattern by a uniform state at  $\epsilon > \epsilon_m$ . The stability interval for a finite fragment of patterns sandwiched between semi-infinite regions of a uniform state slightly depends on the length of that fragment [79]. Note that the noise activates the transition for a metastable state to a truly stable, energetically preferred, state [79].

The coexistence of patterns and uniform states has been revealed for many pattern-forming systems (for a review, see [85]).

## 4. Coarsening in two and three dimensions: curvature effects

### 4.1. Phase separation

First, let us consider domain coarsening for spatially uniform states in a system without the conservation law.

In a two-dimensional (three-dimensional) potential system, the Lyapunov functional can be diminished without annihilation of a domain wall, just by diminishing its length (area). Let us consider a two-dimensional Allen-Cahn equation,

$$\phi_t = \phi_{xx} + \phi_{yy} + \phi - \phi^3 + h, \quad (49)$$

and present the isoline  $\phi(x, y, t) = 0$  (“a front”), which describes the center of a curved domain walls between the stable uniform phases, in the form  $y = H(x, t)$ :  $\phi(x, y, t) < 0$  as  $y < H(x, t)$ ,  $\phi(x, y, t) > 0$  as  $y > H(x, t)$ . One can show that the motion of the domain wall is determined, in the limit of small  $h$  and a small curvature of the front, by the equation [86], [26],

$$\frac{H_t}{\sqrt{1 + H_x^2}} = \frac{H_{xx}}{(1 + H_x^2)^{3/2}} - \frac{3\sqrt{2}}{2} h, \text{ or } v = \kappa - \frac{3\sqrt{2}}{2} h, \quad (50)$$

where  $v$  is the normal velocity, and  $\kappa$  is the curvature of the front. Specifically, in the case  $h = 0$  (both phases have the same free energy), we get just the relation  $v = \kappa$ , which is called *curvature flow*.

As an example, let us consider a round droplet of the phase  $\phi_-$  in the infinite sea of the phase  $\phi_+$ . Because  $\kappa = 1/R$ , in the case  $h = 0$  the droplet radius is changed according to the law

$$R^2(t) = R(0)^2 - 2t.$$

The droplet collapses during the finite time  $t_* \approx R_0^2/2$ . The obtained life time of the droplet shows that the characteristic coarsening scaling is  $l \sim O(t^{1/2})$ , which is significantly faster than in the one-dimensional case. The same coarsening law is obtained in the 3D case.

Moreover, even for the fractional Allen-Cahn equation,

$$\phi_t = -(-\nabla^2)^{\gamma/2} \phi + \phi - \phi^3 + h, \quad 1 < \gamma < 2, \quad (51)$$

the front motion is determined by a formula similar to (50) (up to numerical coefficients that depend on  $\gamma$ ), and the scaling  $l \sim O(t^{1/2})$  is established on the late stage of coarsening (at the initial state,  $l \sim O(t^{1/\gamma})$ , according to the scaling properties of the linearized equation) [37].

A more significant change of the front dynamics is produced by *memory*, when the temporal evolution of the order parameter is governed by the equation

$$\phi_t = - \int_0^t a(t-s) \frac{\delta F}{\delta \phi}(x, s) ds. \quad (52)$$

Equation (50) is replaced by

$$\frac{v_t}{1 - \alpha v^2} + \gamma v = \kappa - \frac{3\sqrt{2}}{2} h (1 - \alpha v^2)^{1/2}, \quad (53)$$

where the constants  $\alpha$  and  $\gamma$  are determined by the Laplace transform of the kernel  $a(t-s)$  [87], [88].

In the presence of a conservation law, using the Cahn-Hilliard equation, one can find that the evaporation of a single round droplet of the phase  $\phi_-$  in the infinite sea of the phase  $\phi_+$  is governed by the equation [6]

$$R^3(t) = R^3(0) - \frac{3}{2} \sigma t,$$

where  $\sigma$  is a parameter corresponding to the effective surface tension of the domain wall. Hence, the scaling law  $l \sim O(t^{1/3})$  is predicted. In the case of droplets of different sizes, the main mechanism of coarsening is the growth of big droplets (with a smaller curvature) at the expense of small droplets (with a larger curvature), which leads to “flattening” of the interphase boundary and hence the decrease of the Lyapunov functional (“Ostwald ripening”). Lifshitz and Slyozov [89] and Wagner [90] have developed a kinetic theory of the phase separation in the limit of small concentration of the minority phase. A detailed description of that theory and its extensions can be found in [21]. Here we mention some basic results. There exists a critical radius  $R_c(t) \sim t^{1/3}$  such that smaller droplets evaporate by diffusion, while larger droplets grow by absorbing the matter through the majority phase. They have obtained a self-similar droplet radius distribution and found the law  $R(t) \sim t^{1/3}$  for the characteristic droplet radius. Later, the latter law was confirmed, theoretically and numerically, for arbitrary concentrations of phases [91]–[94]. Note that at shorter time after the beginning of the phase separation process, a scaling law  $R(t) \sim t^{1/4}$  has been predicted and observed [95]–[97]. The same scaling laws are observed in the framework of a more general model,

$$\phi_t = -\nabla \cdot [M(\phi)(\nabla^2 \phi + \phi - \phi^3)] \quad (54)$$

with a non-constant mobility function  $M(\phi)$ . For instance, the crossover from  $R(t) \sim t^{1/4}$  to  $R(t) \sim t^{1/3}$  has been observed for  $M(\phi) = 1 - \phi^2$  [98].

A nontrivial kind of Cahn-Hilliard equation has been derived for the description of flows in thin liquid films in the presence of disjoining pressure [48]–[51]. Here the conservation law is the the conservation of the liquid volume, while “the phases” are a macroscopic film and a mesoscopic “precursor film”. The Cahn-Hilliard equation describes the decomposition of a film into droplets connected by the thin precursor film. The coarsening of droplets is due to the growth of large droplets at the expense of small ones and because of the motion of droplets leading to their coalescence. In the framework of the standard model [49], the coarsening law is  $N(t) \sim t^{-2/5}$ , where  $N(t)$  is the number of droplets. Generalizations of that model leading to different coarsening rates can be found in [52].

A specific kind of coarsening takes place if there are more than two thermodynamically equivalent phases [99], [100], e.g., because of different possible orientations of the spin (the list of examples can be found in [100]). The problem can be modelled by means of an *overdamped sine-Gordon equation* similar the Allen-Cahn equation (7) but with the potential  $V(\phi) = \cos p\phi - 1$ ; the stable equilibrium phases correspond to the potential maxima,  $\phi = 2\pi m/p$ ,  $m = 0, 1, \dots, p-1$ . For  $p = 2$ , the coarsening is similar to that for the Allen-Cahn equation in any dimension. If

$p > 2$ , the coarsening in 1D is determined by the exponentially weak interaction of domain walls, which can be now either attractive or repulsive. For  $p \geq d + 1$ , a logarithmic rate of coarsening is also predicted [100].

Coarsening in non-potential systems was studied using isotropic [62] and anisotropic [59] generalizations of the convective Cahn-Hilliard equation.

A numerical analysis of coarsening versus pattern formation in non-potential pattern-forming systems has been carried out in [101].

#### 4.2. Pattern ordering

The naturally appearing patterns usually contain numerous defects [102]. Specifically, the pattern may have a multidomain structure. One can distinguish between two kinds of domain walls in periodic patterns [41]. The first kind of domain walls separates patterns of different symmetry, which are generally not energetically equivalent (e.g., stripe patterns and hexagonal patterns). In that case, the domain wall tends to move expanding the energetically preferred domain, but it can be stopped by pinning, as it was explained in the previous sections [41]. The second kind of domain walls separates patterns of the same type but with different orientations or different values of the wavelength. Domains of different orientations appear spontaneously or are created by the side walls in a finite region [103]. Different scenarios of domain wall evolution are possible [41]: (i) the domain wall can be a source of a wavenumber selection, similarly to a side wall [104] or a ramp smoothly matching pattern region with a subcritical region [105]; (ii) the domain wall can be destroyed by an intrinsic instability; (iii) it can spread and smooth down.

Besides domain walls, patterns contain dislocations [106], [107], coupled pairs of dislocations [108], [109], and disclinations [110] - [112]. Their motion is also a significant factor of the ordering in periodic patterns [41], [109].

While some specific phenomena related to the dynamics of defects in patterns have been a subject of a theoretical analysis, the full picture of pattern ordering is studied mostly by means of numerical simulations of dynamical equations (possibly with noise) or experimentally.

In isotropic systems with symmetry  $\phi \rightarrow -\phi$  (e.g., for (2) or (47)), stripes of different orientations are generated. That allows a simplified description of patterns by the phase field  $\phi(\mathbf{x}, t)$ ,  $\phi(\mathbf{x}, t) = \phi_0 \cos[\psi(\mathbf{x}, t)]$ , with the local wavevector  $\mathbf{k}(\mathbf{x}, t) = \nabla\psi(\mathbf{x}, t)$  [113]. The numerical simulations reveal power laws for the growth of domains and elimination of domain walls, dislocations and disclinations for the original equation (47) [114] and for the phase equation [115]. Computations carried out for (2) show that on the background of the final state, which is a unidomain structure, the orientational two-time correlation function has a power-law asymptotics, while the spatial two-point correlation function is subject to a transition from a power law to an exponential law with time [116].

If the symmetry  $\phi \rightarrow -\phi$  is violated (due to an external field [117] or a cubic term in the free energy density [118]), a competition between stripes and hexagons takes place. As an example, let us mention ordering in patterns governed by a generalized Swift-Hohenberg equation

$$\phi_t = -\frac{\delta F}{\delta \phi}, \quad F\{\phi\} = \int \left\{ -\frac{\epsilon}{2}\phi^2 + \frac{1}{4}\phi^4 + \frac{s}{3}\phi^3 + \frac{1}{2}[(\nabla^2 + 1)\phi]^2 \right\} d\mathbf{x}, \quad \mathbf{x} = (x, y). \quad (55)$$

Changing  $s$ , one can arrange a transition between stripes and hexagons and vice versa. The analysis of the transition has been done [118] by studying the structure factor  $S(\mathbf{k}, t) = \langle |\hat{\phi}(\mathbf{k}, t)|^2 \rangle$ , where  $\hat{\phi}(\mathbf{k}, t)$  is the Fourier transform of the order parameter  $\phi(\mathbf{x}, t)$ , and  $\langle \rangle$  denotes ensemble averaging. One has found different scaling laws for ordering the stripes, for the growth of hexagonal domains due to the stripe-to-hexagon transition, and for the growth of stripes from a disordered hexagonal patterns.

Oriental ordering in hexagonal patterns has been studied experimentally in [119] and numerically in [120], using the modification of equation (2) with broken inversion theory. Note that the problem of the orientational ordering is related to the problem of coarsening in the system with degenerate phases studied in [99], [100].

In conclusion, we have reviewed basic models and effects related to coarsening in pattern forming systems. Recently, investigations of more complex cases have been initiated, e.g., domain coarsening in an oscillatory patterns [121], pattern coarsening in time-dependent domains [122], and patterns in networks [123]. These subjects are beyond the scope of the present review.

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